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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 7 : <b>C01B 3/58</b>	<b>A1</b>	(11) International Publication Number: <b>WO 00/17097</b> (43) International Publication Date: 30 March 2000 (30.03.00)
<p>(21) International Application Number: PCT/NL99/00584</p> <p>(22) International Filing Date: 21 September 1999 (21.09.99)</p> <p>(30) Priority Data: 1010140 21 September 1998 (21.09.98) NL</p> <p>(71) Applicant (for all designated States except US): STICHTING ENERGIEONDERZOEK CENTRUM NEDERLAND [NL/NL]; Westerduinweg 3, NL-1755 LE Petten (NL).</p> <p>(72) Inventors; and (75) Inventors/Applicants (for US only): DE WILD, Paulus, Johannes [NL/NL]; Leeststraat 13, NL-1825 JL Alkmaar (NL), VERHAAK, Michael, Johannes, Franciscus, Maria [NL/NL]; Westerweg 41, NL-1815 DB Alkmaar (NL), BAKKER, Dianna, Fokelina [NL/NL]; Vermeerstraat 16, NL-1761 WR Anna Paulowna (NL).</p> <p>(74) Agent: DE BRUIJN, Leendert, C.; Nederlandsch Octrooibureau, Scheveningseweg 82, P.O. Box 29720, NL-2502 LS The Hague (NL).</p>	<p>(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).</p> <p><b>Published</b> With international search report. In English translation (filed in Dutch).</p>	
<p>(54) Title: CATALYSTS FOR THE SELECTIVE OXIDATION OF CARBON MONOXIDE IN HYDROGEN-CONTAINING GASES</p>		
<p>(57) Abstract</p> <p>The invention relates to a method for the selective catalytic oxidation of carbonmonoxide (CO) in H<sub>2</sub>-rich, CO<sub>2</sub>- and H<sub>2</sub>-containing gases in the presence of a noble metal catalyst on an alumina carrier with the addition of air as oxidising agent. According to the invention, with this method <math>\alpha</math>-Al<sub>2</sub>O<sub>3</sub> is used as carrier material. The specific surface area of the <math>\alpha</math>-Al<sub>2</sub>O<sub>3</sub> carrier can be less than 25 m<sup>2</sup>/gram. The noble metal can be ruthenium or platinum and preferably a combination of ruthenium (Ru) and platinum (Pt), the sum of the quantities of Ru and Pt being less than or equal to 1.0 % (m/m).</p>		

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## Catalysts for the selective oxidation of carbon monoxide in hydrogen-containing gases

### Introduction/background information

The invention relates to a method for the selective catalytic oxidation of carbon  
5 monoxide (CO) in the presence of a noble metal catalyst on an alumina carrier.

Fuel cells are being investigated in many places as a possible energy source for driving  
vehicles and for stationary generation of electricity. The use of fuel cells is still highly  
dependent on the availability of the fuel: hydrogen (H<sub>2</sub>). It is not to be expected that an  
infrastructure for hydrogen will be set up within the foreseeable future. Especially for mobile  
10 applications, it is therefore necessary to transport an available fuel, or a fuel that becomes  
available, and to convert this to hydrogen as the feed for the fuel cell.

A gas mixture that consists mainly of hydrogen and carbon dioxide (CO<sub>2</sub>) is then  
produced - for example via steam reforming and/or partial oxidation - from fuels such as  
methane, LPG, methanol, petrol, diesel and other hydrocarbons. Said gas mixture, which is  
15 rich in hydrogen, is then fed to the fuel cell which generates electricity by an electrochemical  
reaction of hydrogen with oxygen.

However, a certain amount of carbon monoxide (CO) is also always liberated during  
the conversion of said fuels into hydrogen. For instance, a gas mixture of, for example,  
75 % (V/V) H<sub>2</sub>, 24 % (V/V) CO<sub>2</sub> and 1 % (V/V) CO is produced on steam reforming of  
20 methanol. A solid polymer fuel cell, the major candidate for transport applications, is  
extremely sensitive to CO, which even in low concentrations (0.01 % (V/V)) has an adverse  
effect on the performance of the fuel cell. For a usable system it is therefore necessary to  
remove CO down to the said level and preferably down to a lower level (< 0.005 % (V/V),  
50 ppm). A technically attractive option for removing CO from H<sub>2</sub>-containing gas streams is  
25 by means of selective oxidation of CO to CO<sub>2</sub> at low temperature (100 °C - 200 °C). In this  
context it is important that the consumption of hydrogen by non-selective oxidation to water  
is minimised.

The power of ruthenium (Ru) to catalyse the oxidation of CO is, for example, known  
from the ammonia synthesis process. Thus, it is known from US Patent 3 216 782 (9  
30 November 1965) that 0.5 % (m/m) Ru on alumina (Al<sub>2</sub>O<sub>3</sub>) is capable of oxidising  
0.055 - 0.6 % (V/V) CO in the presence of H<sub>2</sub> at between 120 °C and 160 °C to a level of less  
than 15 ppm. In this case it is necessary that the quantity of oxygen (O<sub>2</sub>) added is such that  
the molar O<sub>2</sub>/CO ratio is between 1 and 2. The excess oxygen which is not needed for the

oxidation of CO reacts with hydrogen to give water. It has not been investigated whether this Ru catalyst is also capable of oxidising CO from a typical reformat gas to a CO level of 15 ppm under the same conditions (temperature, O<sub>2</sub>/CO ratio).

In the Journal of Catalysis 142 (1993), Academic Press Inc., pages 257-259, S.H. Oh and R.M. Sinkevitch describe 0.5 % (m/m) Ru/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> as highly effective in the complete oxidation, at low temperature (100 °C), of 900 ppm CO with 800 ppm oxygen (O<sub>2</sub>) in a gas mixture which also contains 0.85 % (V/V) H<sub>2</sub>, with the remainder being N<sub>2</sub>. Data on the stability of the Ru catalyst are not given in the article and in addition the behaviour of the catalyst in a realistic reformat gas containing H<sub>2</sub>, CO<sub>2</sub>, H<sub>2</sub>O and CO in much higher concentrations was not investigated.

#### Current state of the art

European Patent EP 0 743 694 A1 (20 November 1996) refers to an oxidation unit for the selective oxidation of CO in H<sub>2</sub>-rich gas at a reaction temperature of between 80 °C and 100 °C. A molar ratio of O<sub>2</sub>/CO of 3 is used. The final CO content is a few ppm. The excess oxygen reacts with hydrogen to give water. The catalyst consists of a 0.2 % (m/m) - 0.5 % (m/m) Pt-Ru alloy on Al<sub>2</sub>O<sub>3</sub>. No examples which would show the stability of the catalyst are given.

US Patent 5 674 460 (7 October 1997) describes a structured reactor for the catalytic removal of CO from H<sub>2</sub>-rich gas at between 90 °C and 230 °C. Depending on the temperature, the catalyst in this case consists of Pt on  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>, Pt on zeolite-Y or Ru on  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>. The invention is explained solely on the basis of 5 % (m/m) Pt on  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>, by means of which the CO content can be reduced to about 40 ppm at a reaction temperature of between 80 °C and 130 °C. No stability data are given in this patent either.

In the Journal of Catalysis 168 (1997), Academic Press, pages 125-127, R.M. Torres Sanchez et al. describe gold on manganese oxide as an alternative catalyst for the oxidation of CO in H<sub>2</sub> at low temperatures (approximately 50 °C). In particular the price, due to the high gold loading (approximately 4 - 10 % (m/m)), makes the use of this type of catalyst less interesting. Moreover, this type of catalyst is able to withstand carbon dioxide to only a limited extent.

It is not clear from the above whether the catalysts of the prior art are suitable for the selective oxidation of CO in H<sub>2</sub>-rich reformat gas mixtures where there is high activity in conjunction with good stability in the temperature range 100 °C - 200 °C and where a low

oxygen excess can be used to minimise the hydrogen consumption.

#### Discovery of new catalyst

One aim of the present invention is to provide a method for the selective catalytic  
5 oxidation of CO from H<sub>2</sub>-rich, CO<sub>2</sub>- and H<sub>2</sub>O-containing (reformat) gas mixtures, making  
use of as small as possible an amount of oxygen and at relatively low temperature. A further  
aim of the present invention is to provide a catalyst which has high chemical and thermal  
stability and can be produced in a cost-effective manner by means of a simple method of  
preparation from commercially available starting materials and a low noble metal loading.

10 The use of commercially available  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> as carrier material in the preparation of  
0.5 % (m/m) Ru on Al<sub>2</sub>O<sub>3</sub> led, surprisingly, to a catalyst which in the temperature range  
120 °C - 160 °C combines high activity (> 99% conversion of CO) with high stability (a CO  
conversion of at least 97 % for a period of at least 50 hours) in the oxidation of CO with a  
relatively small excess of oxygen in dilute reformat gas. These results were found to be  
15 appreciably better than the results which were obtained with a commercially available  
0.5 % (m/m) ruthenium catalyst with  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> as the carrier (specific surface area > 100 m<sup>2</sup>/g),  
which is representative of the catalysts used in the abovementioned studies and reflects the  
prior art.

It has also been found that the addition of Pt and the lowering of the total noble metal  
20 loading resulted in a catalyst which showed even better stability for the selective oxidation of  
CO in both dilute and undiluted reformat gas (a CO conversion of at least 99 % for a period  
of at least 50 hours).

It has furthermore been found that in particular the nature and the specific surface area  
of the Al<sub>2</sub>O<sub>3</sub> carrier used are the factors determining the exceptional performance of the Ru  
25 and Ru-Pt catalysts according to the present invention. Preferably, alumina is used in the form  
of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>. A highly active and stable catalyst is formed when the specific surface area of the  
 $\alpha$ -Al<sub>2</sub>O<sub>3</sub> is in the range from 3 m<sup>2</sup>/g to 25 m<sup>2</sup>/g.

The catalysts in the present invention can be prepared in a simple manner via a standard  
impregnation method from commercially available starting materials. Compared with the  
30 current state of the art, the method according to the present invention has the following  
advantages:

- complete oxidation of CO to CO<sub>2</sub> in the temperature range 120 °C to 160 °C with only a  
small excess of oxygen (O<sub>2</sub>/CO = 1) compared with the stoichiometrically required quantity

of oxygen ( $O_2/CO = 0.5$ ),

- minimal hydrogen consumption as a result of low oxygen excess ( $O_2/CO = 1$ ),
- stable action at 130 °C in simulated reformat gas (0.5 % (V/V) CO, 0.5 % (V/V)  $O_2$ , 74 % (V/V)  $H_2$ , 19 % (V/V)  $CO_2$  and 6 % (V/V)  $H_2O$ ) for a period of at least 50 hours
- 5 (residual quantity of CO < 50 ppm),
- low noble metal loading of less than 0.5 % (m/m).

$\alpha-Al_2O_3$  is a commercial product that is used, inter alia, in the electronics industry in the production of thick and thin substrate layers by tape casting. Another application is the production of industrial ceramics.

- 10 The use of this  $\alpha-Al_2O_3$  as carrier for a selective oxidation catalyst for CO in  $H_2$ -rich gas mixtures has not been described before.

The invention will be explained in more detail on the basis of the following examples together with the appended figures.

- 15 In the figures:

Figure 1 shows the activity of a 0.5 % (m/m) Ru-on- $\alpha-Al_2O_3$  catalyst (code A1Ru-5) compared with the activity of a commercial Ru catalyst with 0.5 % (m/m) Ru on  $\gamma-Al_2O_3$  (code G1RuC-5) in the oxidation of CO in dilute reformat gas,

- 20 Figure 2 shows the stability of A1Ru-5 in the CO oxidation at 130 °C compared with the stability of G1RuC-5 in dilute reformat gas,

Figure 3 shows the activity of a 0.25 % (m/m) Ru, 0.125 % (m/m) Pt-on- $\alpha-Al_2O_3$  catalyst (code A1RuPt-48) compared with the activity of A1Ru-5 in the oxidation of CO in dilute reformat gas,

- 25 Figure 4 shows the stability of A1RuPt-48 in the CO oxidation at 130 °C compared with the stability of A1Ru-5 in dilute reformat gas,

Figure 5 shows the activity of A1RuPt-48 in the oxidation of CO as a function of the reformat gas composition,

Figure 6 shows the stability of A1RuPt-48 in the CO oxidation at 130 °C as a function of the reformat gas composition and

- 30 Figure 7 shows the activity of A2RuPt-48 compared with the activity of A1Ru-5 and G3Ru-5 in the oxidation of CO in undiluted reformat gas.

In the following tests the Ru-on- $\alpha-Al_2O_3$  and the Ru-Pt-on- $\alpha-Al_2O_3$  catalysts were prepared by impregnation of a commercial  $\alpha-Al_2O_3$  carrier with solutions of the salts

5 ruthenium nitrosyl nitrate and hexachloroplatinic acid. The effect of the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> carrier on the CO oxidation activity and stability of the catalyst is determined under III below. The effect of the addition of Pt and the lowering of the total noble metal loading on the catalyst activity and stability is given under IV. Finally, the activity and the stability of the catalyst as a function of the composition of the reformat gas are determined under V.

#### I. Preparation of Ru-on- $\alpha$ -Al<sub>2</sub>O<sub>3</sub> and Ru-Pt-on- $\alpha$ -Al<sub>2</sub>O<sub>3</sub> catalysts

The catalysts according to the present invention were prepared by dry impregnation of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> powder with solutions of ruthenium nitrosyl nitrate ((Ru(NO)(NO<sub>3</sub>)<sub>x</sub>(OH)<sub>y</sub> (x+y=3),  
10 Ru content of the solution 1.5 % (m/m)) and hexachloroplatinic acid (H<sub>2</sub>PtCl<sub>6</sub>.xH<sub>2</sub>O), Pt content 0.5 % (m/m)).

The 0.5 % (m/m) Ru-on- $\alpha$ -Al<sub>2</sub>O<sub>3</sub> catalyst (code A1Ru-5) was prepared by adding 5 gram of the Ru solution to 15 gram of the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> powder in a glass beaker and then stirring well until a pasty substance was formed. This paste was then dried in air in an oven for 16  
15 hours at 80 °C. During drying the setting paste was stirred several times. After drying, the solid material was finely ground to a homogeneous powder with the aid of a mortar. The powder thus produced was then pressed to give a pill. After crushing the pill in a mortar a 0.25 mm to 0.5 mm sieve fraction was prepared for the catalytic measurements. The catalyst prepared was stored in a polyethylene sample bottle at room temperature.

20 In the case of the 0.25 % (m/m) Ru and 0.125 % (m/m) Pt-on- $\alpha$ -Al<sub>2</sub>O<sub>3</sub> catalysts (codes A1RuPt-48 and A2RuPt-48), first 1.68 gram of the Ru solution and then 2.51 gram of the Pt solution were added to 10 gram of the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> powder. The subsequent preparation steps were identical to those described above for A1Ru-5.

#### 25 II. Test apparatus and test procedure

The conversion of CO was studied in an automated micro-flow set-up operating under atmospheric pressure. The following gases were available to the set-up: N<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>, CO<sub>2</sub>, CO and H<sub>2</sub>O. It was possible to measure the gases H<sub>2</sub>, CO<sub>2</sub> and CO with the aid of a Perkin-Elmer model 8500 gas chromatograph equipped with a methanizer, connected in series, a  
30 TCD and an FID. A pneumatically controlled 6-way tap was used for sampling the product gas. CO was also measured occasionally with an Elsag Bailey Hartmann & Braun model URAS 10E ND-IR analyser.

The precursor was contained in a Pyrex glass reactor having an internal diameter of

10 mm. The catalyst bed was covered with glass wool and a layer of glass beads. The height of the catalyst bed was approximately 5 mm, whilst the gas flow was approximately 75 ml/min. The space velocity (SV) was approximately  $11,000 \text{ h}^{-1}$  in this case. The amount of precursor required (0.25 mm - 0.5 mm fraction) was 200 or 400 mg. The temperature was  
5 measured immediately below the catalyst bed using a CrAl thermocouple.

During the measurements the catalyst sample was exposed to a pre-mixed gas containing 0.5 % (V/V) CO, 0.5 % (V/V) O<sub>2</sub>, 5 or 19 % (V/V) CO<sub>2</sub>, 15, 51 or 74 % (V/V) H<sub>2</sub>, 6 or 7 % (V/V) H<sub>2</sub>O, with the remainder being N<sub>2</sub>. Prior to the CO oxidation measurement the catalyst sample was pre-treated with, successively, air at 400 °C and 25 % (V/V) H<sub>2</sub> in N<sub>2</sub>  
10 at 550 °C for activation. The activated catalyst was then cooled under H<sub>2</sub>/N<sub>2</sub> to the starting temperature for the test. The reactor was flushed with N<sub>2</sub> for approximately 10 minutes each time the gas composition was changed. For activity measurements the starting temperature was always 80 °C, after which the reactor temperature was raised in 10 °C steps to a final temperature of 250 °C. The CO conversion was determined at each temperature. For stability  
15 measurements the catalyst bed was first brought to the measurement temperature under H<sub>2</sub>/N<sub>2</sub> after the pretreatment, after which the CO conversion was determined once an hour for a period of 50 hours. The general test conditions for the CO oxidation measurements are given in Table 1.

The CO conversion was calculated on the basis of the amount of CO in the product gas  
20 (CO<sub>out</sub>) using the GC and the amount of CO in the feed gas (CO<sub>in</sub> = 0.5 % (V/V)) determined using the GC in accordance with:  $\text{CO conversion (in \%)} = 100 + (\text{CO}_{\text{in}} - \text{CO}_{\text{out}})/\text{CO}_{\text{in}}$ . Using the NDIR it was separately determined that the detection limit of the GC for CO was approximately 25-30 ppm.



**Table 1 General test conditions**

Weight of catalyst sample	200-400 mg
Volume of catalyst bed	approx. 0.4 - 0.6 ml
Particle size	0.25-0.50 mm
Gas flow rate	75 ml/min
Spatial velocity of the gas per hour (GHSV)	10,000 - 15,000 h <sup>-1</sup>
Feed gases	
Reformate gas 1	0.5% CO, 0.5% O <sub>2</sub> , 15% H <sub>2</sub> , 5% CO <sub>2</sub> , 7% H <sub>2</sub> O, remainder N <sub>2</sub>
Reformate gas 2	0.5% CO, 0.5% O <sub>2</sub> , 51% H <sub>2</sub> , 5% CO <sub>2</sub> , 7% H <sub>2</sub> O, remainder N <sub>2</sub>
Reformate gas 3	0.5% CO, 0.5% O <sub>2</sub> , 74% H <sub>2</sub> , 19% CO <sub>2</sub> , 6% H <sub>2</sub> O
Total pressure	atmospheric
Temperature of catalyst bed	80 °C - 250 °C (10 °C steps in the activity measurements) 130 °C (stability measurements)

### III. Effect of $\alpha$ -Al<sub>2</sub>O<sub>3</sub> carrier on CO oxidation in dilute reformate gas

The test results for the oxidation of CO with O<sub>2</sub> in dilute reformate gas (gas 1) over the Ru-on-alumina catalyst (code A1Ru-5) show that the use of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> as the carrier for Ru results in both a better activity and a better stability in the oxidation of CO compared with a commercial Ru-on- $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalyst (code G1RuC-5).

Figure 1 shows the activity of A1Ru-5 compared with the activity of G1RuC-5 in the oxidation of CO in dilute reformate gas 1. Catalyst A1Ru-5 achieves a more complete CO conversion over a wider temperature range than does G1RuC-5.

Figure 2 shows the stability in the CO oxidation in dilute reformate gas 1 with A1Ru-5 compared with the stability of G1RuC-5. A1Ru-5 is found to be both more active and more stable in the CO oxidation than G1RuC-5 over a measurement period of 50 hours.

### IV. CO oxidation in dilute reformate gas with Ru-Pt on $\alpha$ -Al<sub>2</sub>O<sub>3</sub>

Test results for the oxidation of CO with O<sub>2</sub> in dilute reformate gas 1 over a Ru-Pt-on- $\alpha$ -Al<sub>2</sub>O<sub>3</sub> catalyst (code A1RuPt-48) demonstrate that the addition of Pt and lowering the total noble metal loading results in a catalyst which is more stable than the A1Ru-5 described above. The addition of platinum and lowering the total noble metal loading was not found to have a significant effect on the activity of the catalyst.

Figure 3 shows the activity of A1RuPt-48, which has a low loading, compared with the activity of A1Ru-5 in the oxidation of CO in dilute reformat gas 1. It can clearly be seen that from 120 °C A1RuPt-48 shows virtually the same CO conversion as a function of the temperature as A1Ru-5. This is despite the lower noble metal loading of A1RuPt-48 compared with A1Ru-5.

Figure 4 shows the stability of A1RuPt-48 in the CO oxidation at 130 °C compared with the stability of A1Ru-5 in dilute reformat gas 1. A1RuPt-48 displays a higher conversion of CO than A1Ru-5 over the entire measurement period.

#### 10 V. Effect of reformat gas composition on CO oxidation with Ru-Pt on $\alpha$ -Al<sub>2</sub>O<sub>3</sub>

Test results for the oxidation of CO with O<sub>2</sub> in various reformat gases 1, 2 and 3 over the Ru-Pt-on- $\alpha$ -Al<sub>2</sub>O<sub>3</sub> catalyst described above show that activity and stability are virtually independent of the composition of the reformat gas.

Figure 5 shows the activity of A1RuPt-48 in the oxidation of CO measured in various reformat gas compositions. Only at the highest temperatures is the conversion of CO in the less dilute reformat gases 2 and 3 somewhat lower than the conversion in the most dilute reformat gas 1.

Figure 6 shows the stability of A1RuPt-48 in the CO oxidation in the three different reformat gases 1, 2 and 3. The very high CO conversion with this catalyst is dependent to only a very slight extent on the composition of the reformat gas; even with simulated undiluted reformat gas 3 there is more than 99% CO conversion over the entire measurement period (residual quantity of CO < 50 ppm).

Figure 7 shows the activity of three catalysts in the oxidation of CO in undiluted reformat gas 3. The various curves in Figure 7 for 0.5 % (m/m) Ru on  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> (code G3Ru-5), 0.5 % (m/m) Ru on  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (code A1Ru-5) and 0.25 % (m/m) Ru and 0.125 % (m/m) Pt on  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (code A2RuPt-48) show the substantial effect of the type of carrier material ( $\gamma$  compared with  $\alpha$ ) and the metal composition (Ru compared with Ru/Pt).

## Claims

1. Method for the selective catalytic oxidation of carbon monoxide (CO) in H<sub>2</sub>-rich, CO<sub>2</sub>- and H<sub>2</sub>O-containing gases in the presence of a noble metal catalyst on an alumina carrier with  
5 the addition of air as oxidising agent, characterised in that  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> is used as carrier material.
2. Method according to Claim 1, characterised in that the specific surface area of the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> carrier is less than 25 m<sup>2</sup>/gram.
- 10 3. Method according to Claim 1, characterised in that the noble metal is ruthenium or platinum and preferably a combination of ruthenium (Ru) and platinum (Pt).
4. Method according to Claims 1 and 3, characterised in that the sum of the quantities of Ru and Pt is less than or equal to 1.0 % (m/m).
- 15 5. Method according to Claim 1, characterised in that the quantity of air added to the gas stream to be treated corresponds to a quantity of oxygen which in molar terms is at most five times as great as the quantity of CO to be removed and preferably is equal to the quantity of CO to be removed and is at least equal to half the quantity of CO to be removed.
- 20 6. Method according to Claim 1, characterised in that the oxidation takes place at a temperature of between 100 °C and 200 °C, preferably of between 120 °C and 180 °C.
- 25 7. Method according to Claims 1 and 6, characterised in that the temperature at which the oxidation takes place is maintained by using the catalyst in or on a reactor which has the features of a heat exchanger.

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fig-1

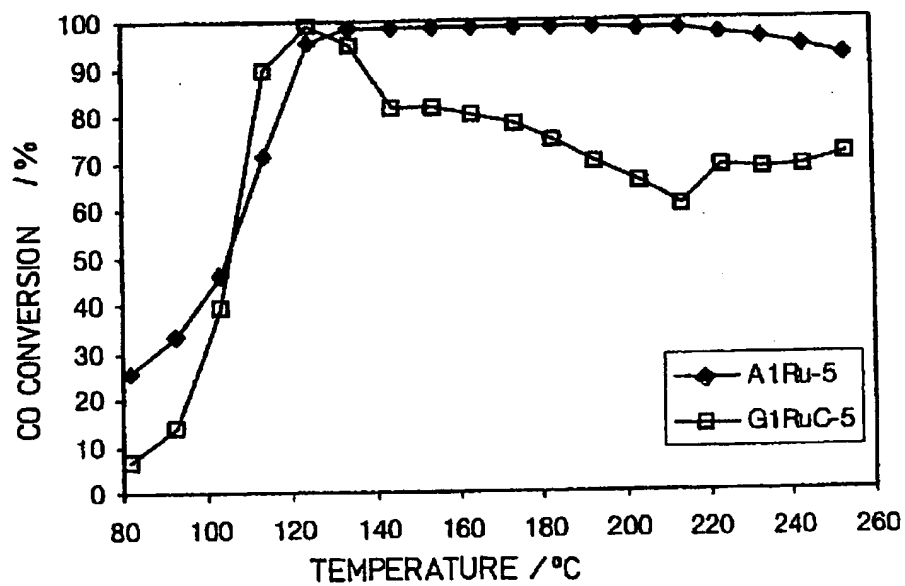
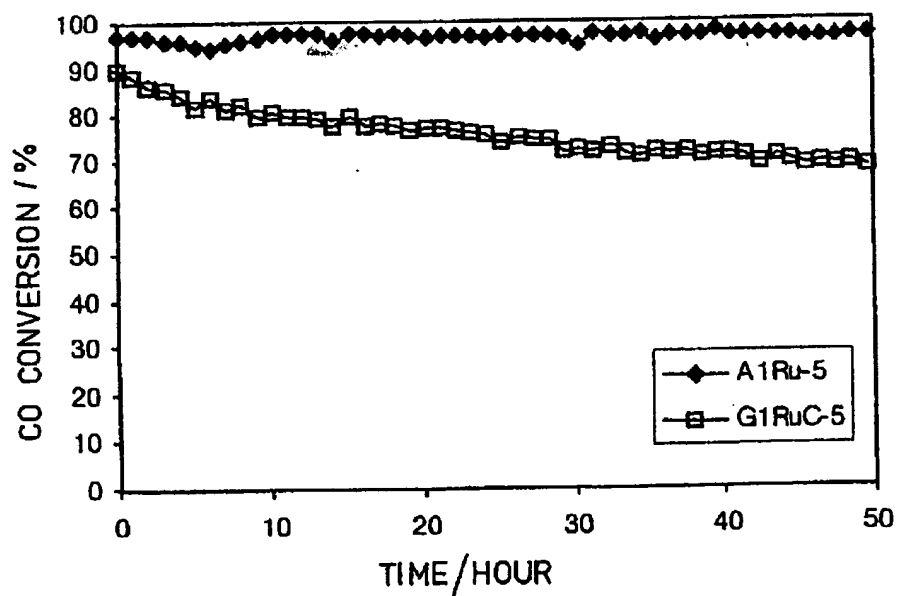


fig-2



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fig-3

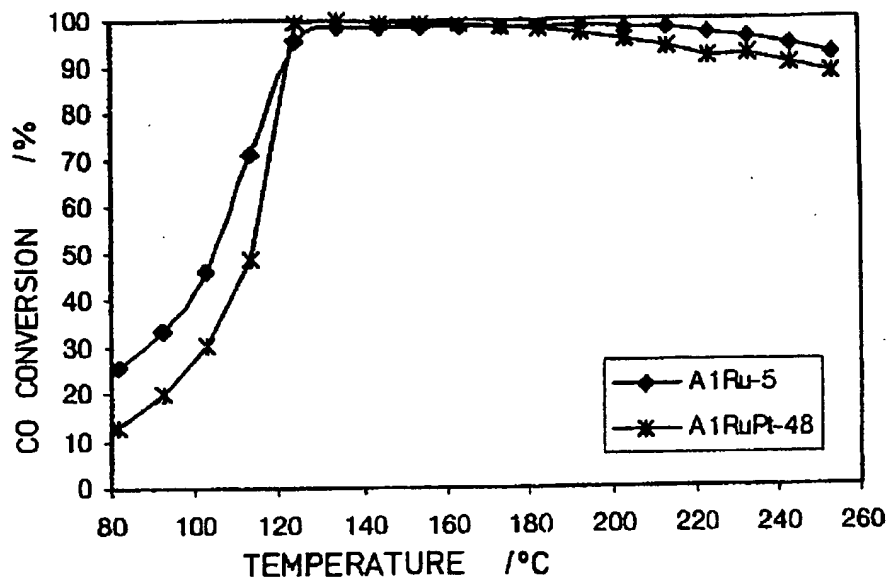
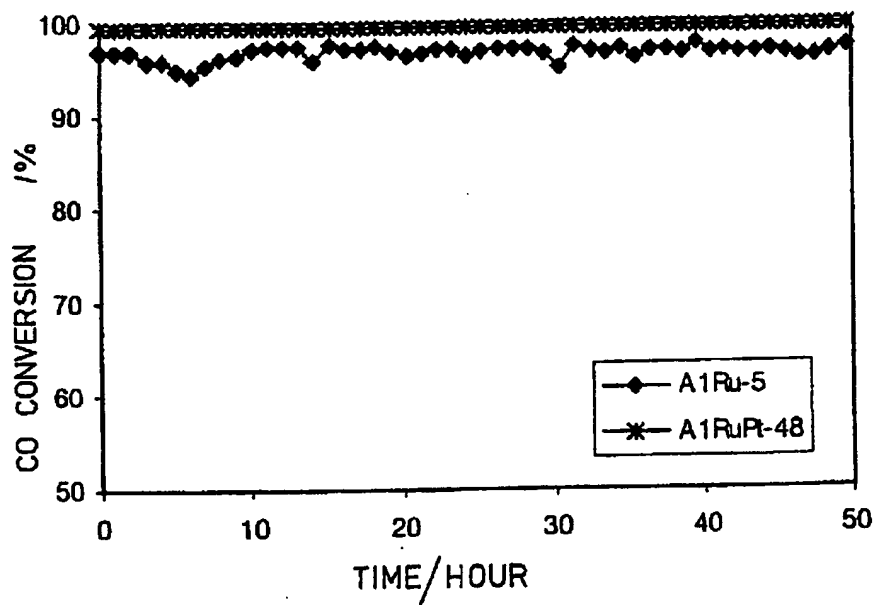


fig-4



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fig- 5

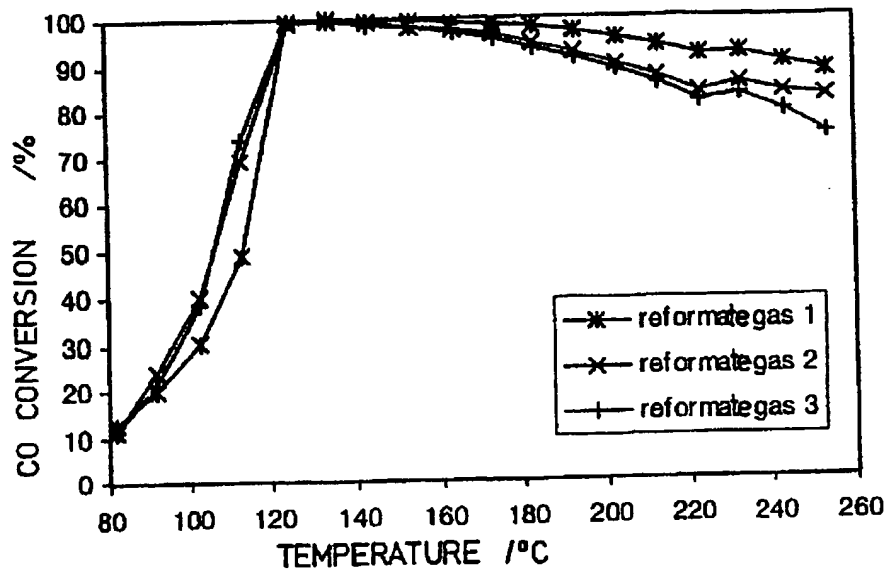
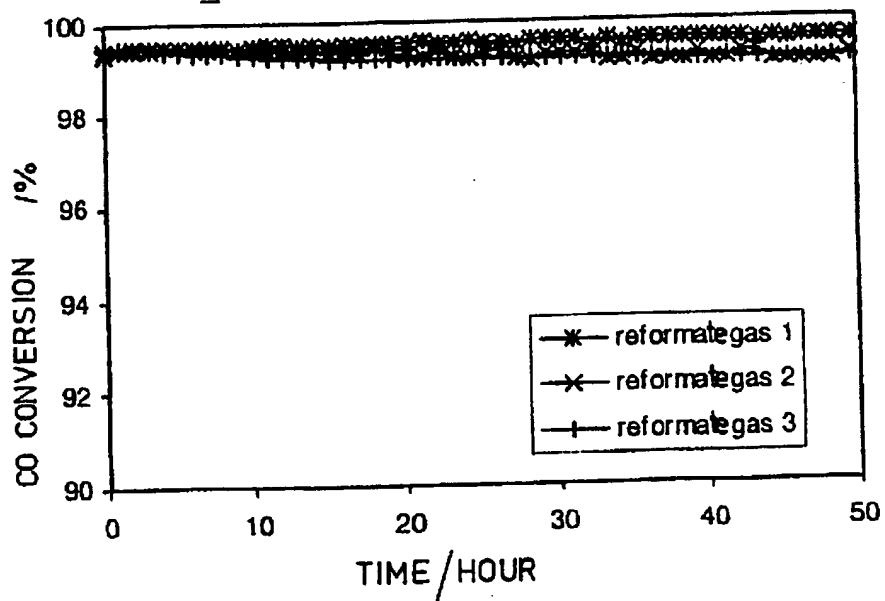
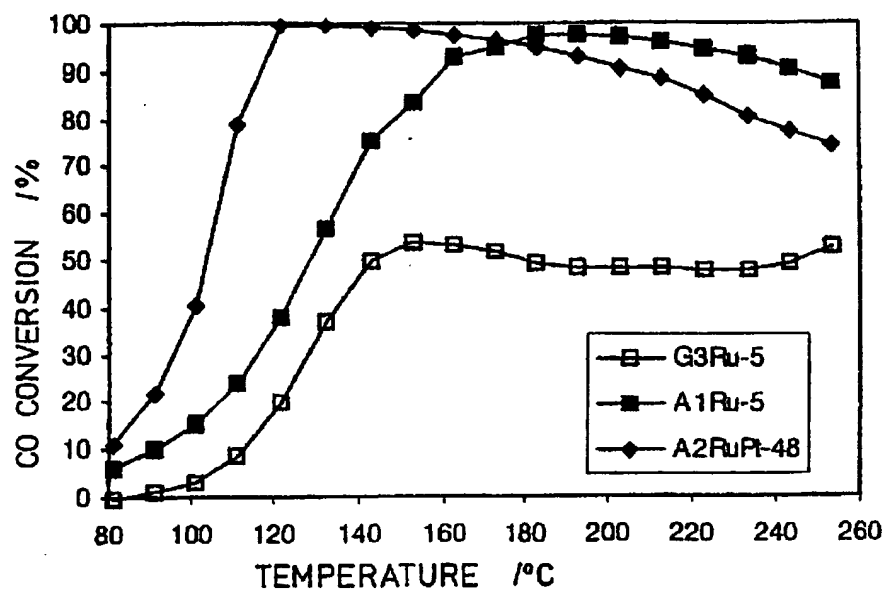


fig- 6



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fig- 7



# INTERNATIONAL SEARCH REPORT

Inter: 1st Application No  
PCT/NL 99/00584

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> IPC 7 C01B3/58		
According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b> Minimum documentation searched (classification system followed by classification symbols) IPC 7 C01B 801D		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practical, search terms used)		
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE 43 34 983 A (DAIMLER-BENZ) 20 April 1995 (1995-04-20) claims	1-7
A	US 5 491 120 A (KENNETH E. VOS ET AL.) 13 February 1996 (1996-02-13) column 2, line 16 -column 3, line 42; claims	1-7
<input type="checkbox"/> Further documents are listed in the continuation of box C. <input checked="" type="checkbox"/> Patent family members are listed in annex.		
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Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentstein 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016		Authorized officer  <div style="text-align: center;">Bogaerts, M</div>



# INTERNATIONAL SEARCH REPORT

information on patent family members

Inter. nal Application No

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